Experimental report

26/01/2022

Proposal: **5-31-2800** **Council:** 4/2020

Title: Magnetic phase diagram under pressure of Ho and Nd pyrochlore iridates

Research area: Physics

This proposal is a new proposal

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Samples: Nd2Ir2O7
Ho2Ir2O7

Instrument	Requested days	Allocated days	From	To
D20	6	3	12/02/2021	15/02/2021

Abstract:

In pyrochlore iridates (formula R2Ir2O7), the iridium magnetic moments apply a molecular field on the rare-earth ions, resulting in a wide variety of magnetic phases. Recently, a theoretical phase diagram was computed, showing that by varying the Ir molecular field and the rare-earth interactions, one can go from an antiferromagnetic state to a fragmented phase (where the magnetic moment fragments into an ordered antiferromagnetic phase and a fluctuating phase) and to a spin ice state. We propose to explore this phase diagram experimentally by measuring the magnetic structure under pressure in Ho2Ir2O7, where we have shown the existence of a fragmented phase, and in Nd2Ir2O7, where the published and our preliminary results suggest that the compound is close to a phase boundary.

Magnetic phase diagram under pressure of Ho pyrochlore iridates.
Diffraction in PE cell at D20.

Aim of the experiment: the aim of this experiment was to probe the influence of isostatic pressure on the magnetic ground state of $Ho_2Ir_2O_7$. Our objective was to explore the (P,T) phase diagram experimentally. Indeed, we expect that applying pressure will change the distances between atoms, and so the magnetic exchange paths. Different phases are expected, mainly all-in / all-out (AIAO), monopole crystal (fragmented), spin-ice and perhaps also ordered spin ice.

We thus essentially planned to follow the intensities of the (220) and (113) AIAO Bragg peaks as a function of pressure and temperature. Table 1 gives the list of AIAO Bragg peaks along with their intensities. If a spin S_i is at a site R_i of the lattice, the magnetic structure factor is given by:

$$F = \sum_i S_i \; e^{iQR_i} \tag{0.1}$$

with an intensity:

$$I_M = \sum_{a,b} F_a^* \; (\delta_{ab} - \frac{Q_a Q_b}{Q^2}) \; F_b \tag{0.2}$$

Index	Intensity I_M	Multiplicity ν_i	$2\theta_i$ ($\lambda = 2.4$)
(111)	0	8	23.5
(002)	0	6	27.2
(220)	85.33	12	38.9
(113)	62.06	24	45.9
(222)	0	8	48.1
(331)	35.93	24	61.7
(224)	28.44	24	70.4
(442)	75.85	24	89.8

Table 1: List of AIAO Bragg peaks with index, scattering angle and squared form factor (intensity).

Experimental conditions: The measurements were carried out on a powder sample at D20, from 12/2 to 15/2/2021. We used a PE pressure cell, allowing one to apply pressure up to 10 GPa. The sample was loaded in a TiZr gasket (to minimize the neutron background) and soaked with a few drops of a (deuterated) ethanol-methanol mixture. Two hemispheres were prepared and then glued together by applying a small pressure of about 80 bar. A small lead chip was added in these hemispheres to measure the pressure *in situ*.

D20 was operated with a PG002 monochromator and $\lambda = 2.4$ Å. Thomas Hansen encouraged us to use the following resolution parameters : U=2.384298, V=-2.185067, W=0.699330, X=0, Y =0.010800 along with nprof=7 (pseudo-Voigt with Gaussian-Lorenzian FWHM parametrization) rather that nprof =5. Smaller FWHM are expected and Thomas suggested to refine them slightly.

We could do 4 cooling-heating sequences, at 0, 4, 8 and 10 GPa. We recorded diffractograms at room temperature, at 80 K and then, while cooling, down to the lowest temperature 5 K. To save time, the cryostat was cooled down very quickly from 300 to 80 K using liquid nitrogen, which was then removed and pushed away with dry air. We could also measure at fixed temperatures of 5, 7, 10 and 15 K, with however different counting times and statistics.

The diffractograms were recorded while moving the whole detector bank by three degrees, which allowed to get steps of 0.05 degrees in 2θ. A script written by Thomas allowed us to collect intensities from the same 2θ and to remove spurious points out of the statistics. The counting time for one of these measurements was about 1 hour, and the temperature of these scans was averaged over this time. However, at 15, 10 , 7 and 5 K (and for 4, 8 and 10 GPa), we measured at fixed temperatures. At 5 K, we measured for 3 hours. At ambient pressure, a He leak (from the pressure circuit) prevented us to cool lower than 17 K. The problem was however fixed at larger loads.

Data analysis: The data proves quite difficult to refine with FP because of background issues (which evolves as a function of temperature) but also because the shape of the peaks is difficult to reproduce. Since the magnetic structure has a propagation vector $\mathbf{q} = 0$, the refinements give only poor confidence in the intensities, scale factor and magnetic moments. We pursued an alternative analysis which consists in three steps:

- Fit with FP the 80 K data to basically determine the scale factor V and lattice parameter a. The background is drawn by hand. This fit also provides the lattice parameters of the lead samples (see Table 2 and figure 1).

P	a (Å)	Scale factor V	Lead a_0 (Å)
ambient	10.22420	0.01011	4.93444
4 GPa	10.17159	0.004559973	4.83788
8 GPa	10.12420	0.002898	4.7463
10 GPa	10.10446	0.00237	4.720389

Table 2: Parameters determined from the FP fit at 80 K.

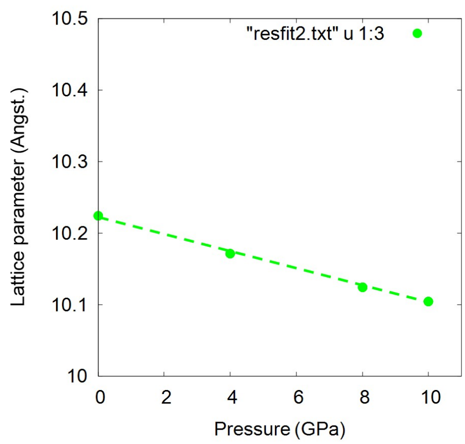

Figure 1: D20 results: $Ho_2Ir_2O_7$ lattice parameter vs pressure at 80 K.

- Calculate the difference between the low temperature and the 80 K data (see Figure 2). Since the lattice parameter does not change much below 80 K, this subtraction eliminates the nuclear signal and we are left with a $\mathbf{q}$-dependent background along with the magnetic signal. Finally, since FP only accounts for a hand-drawn background, we used a different program to fit the differences through the following function:

$$I(\theta) = a + bQ^2 \ + \ A \sum_i \nu_i \ I_i \ e^{-4\ln(2) \ (\frac{\theta-\theta_i}{\delta})^2} \tag{0.3}$$

  A, a, b, δ are the free parameters while the other quantities are taken from Table 1. a and b account for the background, A is the magnetic intensity and δ the FWHM of the magnetic peaks. T

- Renormalize A (or $A \times \delta$) by the scale factor determined at 80 K (A/V) to get rid of the pressure dependent variations. In this way, one determines the relative ratios of the intensities measured at different pressures. To eventually determine the magnetic moments m, it remains to scale the results with a global parameter f, hence the magnetic moments $m = f\ \sqrt{A/V}$. f

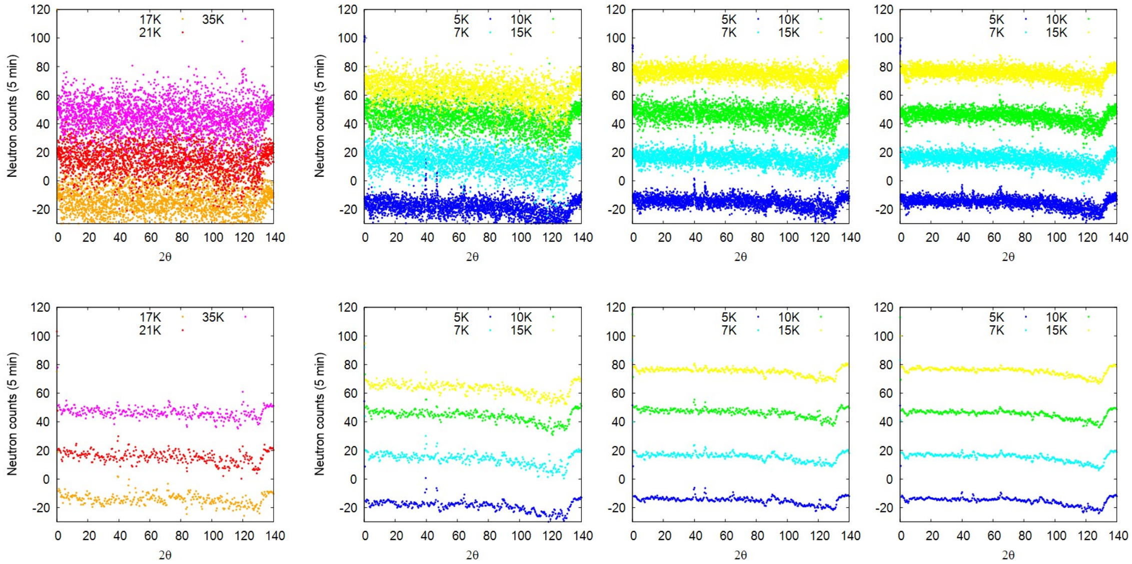

Figure 2: Differences between D20 raw (top) and binned (bottom) data at a given temperature and the 80 K data. From left to right, the panels show ambient, 4, 8 and 10 GPa. The data have been shifted by a fixed quantity to show them on the same graph.

is chosen to reproduce the magnetic moment at ambient pressure, which is known precisely, for instance, at 17 K (see Ref [3]). This fitting procedure yields the evolution sketched in Figure 3. It suggests that high pressures tend to reduce the AIAO ordered phase.

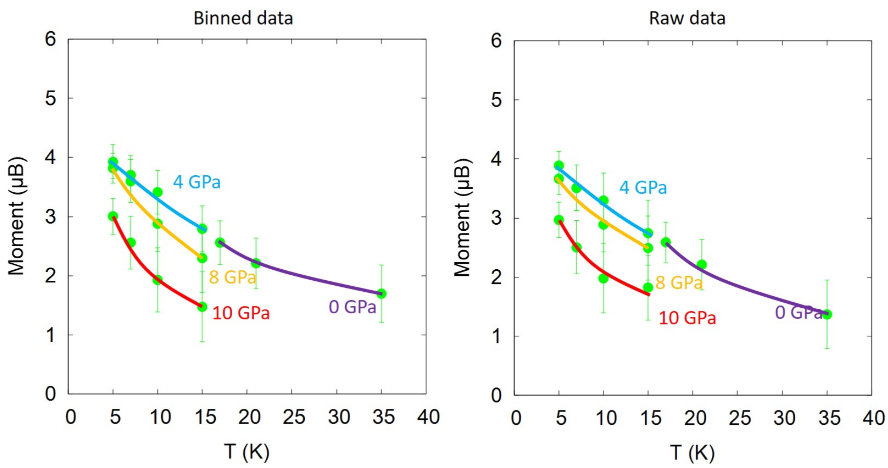

Figure 3: Fitting procedure results. Evolution of the AIAO ordered moment vs T and P (using binned data, left panel, and raw data, right panel).

References:

[1] Wan et al., Phys. Rev. B 83, 205101 (2011).
[2] Lefrançois et al., Phys. Rev. Lett. 114, 247202 (2015).
[3] Lefrançois et al., Phys. Rev. B 99, 060401(R) (2019).
[4] Tomiyasu et al., J. Phys. Soc. Japan 81, 034709 (2012)